REMARKS

The specification has been amended to correct typographical errors by replacing symbols and/or numbers where the "□" appeared. No new matter has been added and the subject matter amended is fully supported by the original specification.

In view of the above, an early action on the merits is now in order and is most respectfully requested.

Respectfully submitted,
BACON & THOMAS, PLLC

By: _____
Eugene Mar
Registration No. 25,893

625 Slaters Lane, Fourth Floor
Alexandria, Virginia 22314
Phone: (703) 683-0500
Facsimile: (703) 683-1080

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same problem since it belongs to this kind of compound. Hence, it is necessary to develop new type of cyanine dye having photostability. In 1999. Shin-ichi Morishima et al., Fuji-film company in Japan, attempted to use the doping method. (Shin-ichi Morishima et al., Jpn. J. Appl. Phys. Vol. 38 (1999), pp.1634-1637). Addition of neutral TCNQ derivative to dye increases the photostability of dye, however, ICNQ is not quite soluble in the common organic solvents so that content of doping dye formulation is limited, therefore, the stability effect is also limited.

SUMMARY OF THE INVENTION

Hence, the aim of this invention is to solve the drawbacks described above. In order to avoid the presence of the drawbacks described above, this invention is to provide a kind of cyanine-TCNQ complex dyes mixture (II, III, and IV), used for the data storage media, with short wavelength (300nm~400nm) and near infrared region (800nm~1000nm), having high absorption for light, resisting the damage of dye from the UV light and singlet oxygen.

The other aim of this invention is to provide a kind of cyanine-TCNQ complex dyes mixture (II, III, and IV), used for the data storage media, having high oxidation potential to prevent the oxidation of dyes.

The other aim of this invention is to provide a kind of cyanine-TCNQ complex dyes mixture (II, III, and IV), used for the data storage media, in which they can adjust the primary absorption position with wavelength in the range of 400~800nm and possess

3H-indole (TCI Chemical) (1.75 g) in benzene was heated at 80℃ ~90 ℃ for 4~8 hours. The solution was evaporated and recrystallized from methanol to give light grey crystals. The yield was 75 %, m.p. 120℃.

(d) Preparation of intermediate (A), the structural formula of (A) as shown in FIG. 6:

A mixed solution of MBTI (6.16 g) and N,N'-diphenylformamidine (1.12 g) in acetic anhydride (20 mL) was heated at 100 ℃ ~ 120 ℃ for 2 hours. After cooling, the reaction mixture was poured into an aqueous solution of sodium perchlorate (1.41 g) to give solid, then was dissolved in dichloromethane, washed with water several times, evaporated and recrystallized from ethanol to give intermediate (A). The yield was 70 %.

(e) Preparation of 2-[3-(1,3-dihyhro-1,1-dimethyl-3-(4'-methoxycar-bonyl)-benzyl-2H-benze[e]indol-2-ylidene)-1-propenyl]1,1-dimethyl-3-butyl-1H indolium TCNQ (II), the structural formula of cyanine TCNQ complex dye (II) as shown in FIG. 2:

Placed 1-(4'-methoxycarbonyl)-2,3,3-trimethyl-4,5-benzo-3H-indole (0.02 mole) and ethyl orthoformate (TCI Chemical) (0.01 mole) in a two-necked flask, dissolved with pyridine, then heated at 110℃~120℃ for 1~3 hours. After cooling, the reaction mixture was poured into an aqueous solution of LiTCNQ (1.24 g) to give solid, recrystallized from methanol to give dark red crystals (III).

The yield was 70 %, m.p. 190 ℃. FIG. 7 shows that the UV/Visible/IR absorption spectrum for cyanine TCNQ complex dye (II), λmax (EtOH) = 568 nm; FIG. 8 and FIG. 9 show that IR spectrum and thermoweighing spectrum(10 ℃/ min) for cyanine TCNQ complex dye (II), respectively.

[Experimental example 2, preparation example]

Preparation of 1-(4'-methoxycarbonyl)benzyl-3,3-trimethyl-1'-(4'-methoxycarbonyl) benzyl-3,3'-dimethylindo-2,2'-trimethine TCNQ (abbreviated as S-TCNQ), S cyanine TCNQ dye, the structural formula of cyanine TCNQ complex dye (III) as shown in FIG. 10, needs through the following described procedure:

(a) Preparation of methyl (4-iodomethyl) benzoate (MIB) first, a mixed solution of 4-chloromethylbenzoyl chloride (TCI Chemical) (1.89g, 0.01mole), methanol (0.32g, 0.01mole), and pyridine (0.791g, 0.01mole) in benzene was heated at 40℃ for 3 hours, then filtered and evaporated to obtain white solids. Dissolved white solids in acetone and added sodium iodide (1.50 g), a mixture was heated at 40 ℃ for 3 hours, filtered, extracted from dichloromethane and water, evaporated to give the light yellow products.

(b) Preparation of 1-(4'-methoxycarbonyl)benzyl-2,3,3-trimethyl-4,5-benzo- 3H-indole:

A mixed solution of MIB (0.01mole) and 2,3,3-trimethyl-4,5-benzo-3H-indole (TCI Chemical) (0.01mole) in MEK was heated at 80℃ for 3 hours. The solution was evaporated and recrystallized

from ethyl acetate to give white crystals.

(c) Preparation of 1-(4'-methoxycarbonyl)benzyl-2,3,3-trimethyl-1'-(4'-methoxycarbonyl) benzyl-3,3'-dimethylindo-2,2'-trimethine TCNQ:

A mixed solution of 1-(4'-methoxycarbonyl)benzyl-2,3,3-trimethyl-4,5-benzo-3H-indole (0.02mole) and ethyl orthoformate (TCI Chemical) (0.01mole) in pyridine was heated at 110℃~120℃ for 1~3 hours. After cooling, the reaction mixture was poured into an aqueous solution of LiTCNQ (1.24g) to give solid, recrystallized from methanol to give dark red crystals (III). The yield was 73%, m.p. 178℃. FIG. 11 shows that the UV/Visible/IR absorption spectrum for cyanine TCNQ complex dye (III), λmax (EtOH) = 551 nm; FIG. 12 and FIG. 13 show that IR spectrum and thermoweighing spectrum (10℃/min) for cyanine TCNQ complex dye (III), respectively.

[Experimental example 3, preparation example]

Preparation of 1-(4''-methoxycarbonyl)benzyl-3,3-dimethyl-1'-(4''-methoxycarbonyl) benzyl-3,3'-dimethylindo-2,2'-pentamethine TCNQ, the structural formula of cyanine TCNQ complex dye (IV) as shown in FIG. 14, needs through the following described procedure:

(a) Preparation of the substituted methyl (4-iodomethyl) benzoate first, then preparation of 1-1-(4'-methoxycarbonyl)benzyl-2,3,3-trimethylindoleninium iodide, finally reaction with 3-anilinoacryl-aldehyde anil gave the structural formula (IV) dye.

(b) Preparation of methyl (4-iodomethyl) benzoate (MIB), the structural formula of MIB as shown in FIG. 3:

A mixed solution of 4-chloromethylbenzoyl chloride (TCI Chemical) (1.89g), methanol (0.32g), and pyridine (0.791g) in benzene (25mL) was heated at 40℃ for 3 hours, filtered and evaporated to obtain white solids. Dissolved white solids in acetone and added sodium iodide (1.50g), a mixture was heated at 40℃ for 3 hours, filtered, extracted from dichloromethane and water, evaporated to give the light yellow solids. The yield was 90%, m.p. 67℃.

(c) Preparation of 1-(4'-methoxycarbonyl)benzyl-2,3,3-trimethyl-4,5-benzo- 3H-indoleninium iodide (MBTI), the structural formula of MBTI as shown in FIG. 5:

A mixed solution of MIB (2.76g) and 2,3,3-trimethyl-4,5-benzo-3H-indolenine (TCI Chemical) (1.75g) in benzene was heated at 80 ℃ ~85 ℃ for 6 hours. The solution was evaporated and recrystallized from ethyl acetate to give light yellow crystals. The yield was 71%, m.p. 109℃.

(d) Preparation of 1-(4''-methoxycarbonyl)benzyl-3,3-dimethyl-1'-(4''-methoxycarbonyl)-benzyl-3',3'-dimethylindo-2,2'-pentamethine TCNQ (IV):

A mixed solution of 1-(4'-methoxycarbonyl)benzyl-2,3,3-trimethylindoleninium iodide (6.16g), 3-anilinoacrylaldehyde anil (TCI Chemical) (2.85g) and sodium acetate (1.64g) in acetic

anhydride (20mL) was heated at 100℃ for 2 hours. After cooling, the reaction mixture was poured into an aqueous solution of LiTCNQ (1.41g) to give solid, which was dissolved in dichloromethane and washed with water several times, evaporated and recrystallized from ethanol to give dark green crystals (IV). The yield was 70%, m.p. 201℃. FIG. 15 shows that the UV/Visible/IR absorption spectrum for cyanine TCNQ complex dye (IV), λmax (EtOH)=649nm; FIG. 16 and FIG. 17 show that IR spectrum and thermoweighing spectrum (10℃/min) for cyanine TCNQ complex dye (IV), respectively.

[Experimental example 4, applied example]

The manufacture process of recordable optical disc:

New type of cyanine TCNQ complex dye (II) (1.50g) and cyanine dye (IV) (0.075g) were dissolved in 2,2,3,3-tetrafluoropropanol to form a 100g solution. The dyes were coated onto blank substrate plate by a spin coater, the whole coating processes of a coater are as follows: a dipping process: at 30~500rpm, 2~10 seconds; a spinning out process: at 1000~3000rpm, 10~30 seconds; and a baking process: at 2000~5000rpm, 10~30 seconds. The recording layer, coated by TCNQ dye onto the substrate plate, has the thickness of about 500 Å ~2000 Å, on which it was then sputtered with a layer of Au, Ag, Al, Cu, Cr and its alloy to form the reflection layer having a thickness of about 500 Å ~1000 Å; finally, combination of a substrate plate sputtered with a reflection layer and a recording layer with the other PC blank substrate plate (e.g. DVD-R 0.6mm